

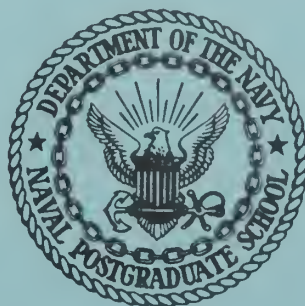
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ANALYSIS OF VOLATILE REACTOR FUEL  
DECOMPOSITION PRODUCTS  
BY SPECTRAL ANALYSIS

Larry Alvin Lindsay

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# UNITED STATES NAVAL POSTGRADUATE SCHOOL



## THESIS

ANALYSIS OF VOLATILE REACTOR FUEL DECOMPOSITION  
PRODUCTS BY SPECTRAL ANALYSIS

by

Larry Alvin Lindsay

June 1968





IDENTIFICATION OF VOLATILE REACTOR FUEL  
DECOMPOSITION PRODUCTS BY SPECTRAL ANALYSIS

by

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LINDSEY, L

ABSTRACT

The volatile radioactive species produced by irradiation of AGN - 201 Reactor fuel elements were obtained by irradiation of the fuel in an evacuated inert container. The following 11 constituent nuclides, including two radioactive daughters, were identified by gamma ray spectrometry:  $I^{131}$ ,  $Xe^{131}$ ,  $Xe^{131m}$ ,  $Xe^{133}$ ,  $Xe^{135}$ ,  $Xe^{138}$ ,  $Kr^{85m}$ ,  $Kr^{87}$ ,  $Kr^{88}$ ,  $Cs^{138}$ , and  $Rb^{88}$ . Additionally, the possible presence of the following species was indicated:  $I^{133}$ ,  $I^{134}$ ,  $I^{135}$ ,  $I^{136}$ ,  $Xe^{137}$ ,  $Kr^{88}$ ,  $K^{89}$ , and  $Rb^{89}$ . The spectra studied were taken with a NaI (Th) crystal and they contained 34 photopeaks. An automated computerized method for half life analysis of complex photopeaks is described.

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## LIST OF DEFINITIONS OF ABBREVIATIONS AND SYMBOLS

1. AGN-201 - The model 201 nuclear reactor produced by Aerojet General Nucleonics Corp., San Bruno, California.
2. A - Atomic mass; or, in Figures 2-5: activity, i.e., disintegrations per channel per unit time.
3.  $A_1, A_2, C_1, C_2, P, W_1, W_2$ : - defined where used.
4. cm - centimeter.
5. Mev - million electron volts.
6. n - neutron.
7. NPS - The Naval Postgraduate School, Monterey, California.
8. SJ - standard joint.
9. SS - stainless steel.
10. ST - standard taper.
11. Z - atomic number, or numbers of protons in the nucleus.
12.  $Z_p$  - defined where used.
13. Elemental symbols:
  - Ba - barium
  - Br - bromine
  - Cs - cesium
  - Cu - copper
  - Hg - mercury
  - I - iodine
  - Kr - krypton
  - La - lanthanum
  - Mg - magnesium
  - Mn - manganese
  - Rb - rubidium
  - Sr - strontium
  - U - uranium
  - Xe - xenon
  - Y - yttrium

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## ANALYSIS OF VOLATILE REACTOR FUEL DECOMPOSITION

### PRODUCTS BY SPECTRAL ANALYSIS

#### I. Introduction

The Naval Postgraduate School Reactor Facility contains an AGN-201 nuclear reactor that has been modified to produce up to 1000 watts for short periods of operation. The core and rods of the reactor contain uranium dioxide suspended uniformly in a polyethylene matrix (1). During operation, of course, numerous fission products are formed, and the polyethylene is radiolytically decomposed by recoil and the conglomerate photon and particulate radiation present. Many of the radioactive substances thus formed are volatile so they must be withdrawn from the reactor in a controlled manner, and this is done routinely. Since the scheme of the disposal is to exhaust these substances into the atmosphere, the characterization of these radioactive substances has considerable practical application. Also, the suspension of the uranium dioxide in polyethylene in the reactor is a system which has, potentially, a very interesting chemistry, because of the multitude of organic and inorganic hot atoms that are mixed intimately together.

Much is known about fission products, per se, and radiation effects on polyethylene, and some research has been done on the gaseous effluents at the NPS reactor. The composition of the gaseous effluents from the NPS reactor has been reported on by Grissom (2), Bredderman (3), and Laskaris (4). The first two investigators assumed that almost all the volume of gas generated was hydrogen. They both calculated the specific activity of the core gas and Bredderman identified six radioactive nuclides by gamma ray spectrometry. Bredderman used the capable, but laborious, method of Heath (5) to determine photopeak half lives. Laskaris used

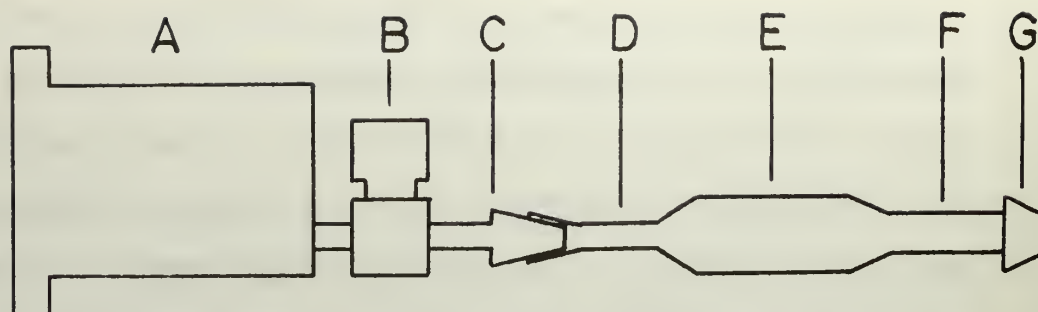
simultaneous gas chromatography and scintillation counting to show that reactor gas was approximately 50% hydrogen by volume, and that xenon and krypton were present.

The previous in house efforts on the analysis of reactor effluents made considerable progress toward explaining the gross characteristics of the situation, however, they were handicapped in several ways. Their samples were taken at the exhaust manifold of the reactor, and this is separated from the reactor core by about 4 meters of 5 mm I.D. copper tubing. It is significant in this regard that the cylindrical core contains, in addition to the fuel, an annular ring of graphite that is about 2.5 centimeters thick, 25 centimeters in diameter, and 25 centimeters high. The fuel - moderator system referred to earlier consists of about 3 kilograms of uranium dioxide and about 12 kilograms of polyethylene. Thus, samples taken at the manifold may not have been truly representative of the gases produced in the core. Reactive species present in only small quantities, for example, iodides and bromides, have a very high probability of being totally absorbed before reaching the sample bottle. Furthermore, every sample was almost certainly different, because it contained an unknown amount of aged gas. The composition of this aged gas was dependent on both the length of time the active constituents had decayed, and the relative rates of diffusion of all the constituents out of the polyethylene mass. Another uncertainty was introduced by the fact that the core is pressurized with water pumped nitrogen which contains greater than atmospheric concentrations of most of the rare gases.

It was believed that the foregoing handicaps were sufficiently important that, if they could be overcome, another attempt at analysis of the core gas was warranted. The problems were circumvented and the analysis was started. The vital factor in meeting the problems was the decision

to obtain fuel gas in a much more controlled manner by irradiation of reactor fuel in an evacuated inert container. This technique changes the project from analysis of the aforementioned core gas to analysis of fuel gas. The principal implications of this are that the samples now analyzed will be void initially of the long lived constituents of the aged gas, but it will be possible to detect many of their short lived precursors. The most important benefits to be derived, however, are that the samples will be reproducible and will contain greater quantities of reactive and trace constituents.

Additionally, the project would afford an excellent opportunity to explore methods of applying the two recently acquired single channel analyzers, with rapid digital read out, to the analysis of complex gamma spectra. It was felt that there was considerable opportunity to contribute to the state of the art of unraveling complex gamma spectra by automating and computerizing data acquisition and reduction. This was taken as a secondary objective of this research.



SAMPLING PROCEDURE: Evacuate entire system to less than 1 micron of Hg; Seal at F and remove G; Irradiate; Pump gas into ampul with liquid nitrogen; Seal at D and remove ampul for counting.

COMPONENTS: A, Al container; B, SS bellows valve; C, SS ST; E, quartz tubing; G, SJ.

Figure 1

## II. Experimental

Reactor fuel elements were irradiated in a hermetically sealed aluminum container in an access part of the reactor, and a sample of the gas pumped with liquid nitrogen directly to a sealed quartz ampul for gamma ray spectrum analysis. Quartz was used to preclude spurious peaks from activation of the ampul. An attempt to activate a sample of quartz stock showed it produced no interfering peaks. Normally one fuel element was irradiated to produce the gas sample, and a typical irradiation was  $10^9$  n/cm<sup>2</sup>/sec for 15 minutes. A fuel element is a circular disc 4 cm in diameter and 1 cm thick that contains 1.0 gram of U<sup>235</sup>. The uranium is present in the form of uranium dioxide that is 20% enriched in U<sup>235</sup>. The sampling procedure is outlined in figure 1 above. The results are described in the sections following this and a description of equipment used is given in Appendix 1.

### III. The Gamma Spectrum of Reactor Fuel Gas

#### A. A Qualitative Analysis

The gamma spectra obtained from the fuel gas have an extremely complex and dynamic nature. The total spectrum contains about thirty-four peaks which can be shown by detailed analysis to represent well over forty gamma rays. The previously reported spectra (3) contained thirteen photopeaks. Illustrative fuel gas spectra are shown in Figures 2, 3, 4, and 5. These spectra are representative of the approximately 400 spectra taken during this project. The number given above a peak in a spectrum corresponds to a peak number in Table I, which contains a list of the corresponding energies. The decade markers on the activity ordinate are useful only for gauging the relative heights of peaks in the same spectrum. The natural background radiation was significant in Spectra L, M, and N, so 80% of the background was subtracted prior to graphing.

Nearly all of the peaks indicate by their shape, shift in peak energy, and complex decay, that they are a composite of several gamma rays. Obvious examples of peaks that show the characteristics mentioned are: the peak at 0.255 Mev which shows two extremely different half lives; the peak(s) that are initially about 0.43 Mev which is apparently at least three peaks of widely different half lives; and the very wide flat-topped peak with a nominal energy of 0.87 must include several gamma rays of slightly different energies and very similar half lives. Only one marked qualitative difference was seen in the spectra obtained from 21 irradiations of the fuel and that was in the 0.255 Mev peak. The height of the peak was much larger in the spectra of one of the runs. There was no readily apparent reason for this phenomenon, and the matter was not pursued. The peak above 3 Mev was seen only in one spectrum, but it was one of the earliest 4 Mev spectra taken during the project. The length of irradiation and neutron flux were varied, and they only affected the amount of gas produced.

# Photopeaks Present in Reactor Fuel Gas Gamma Spectra

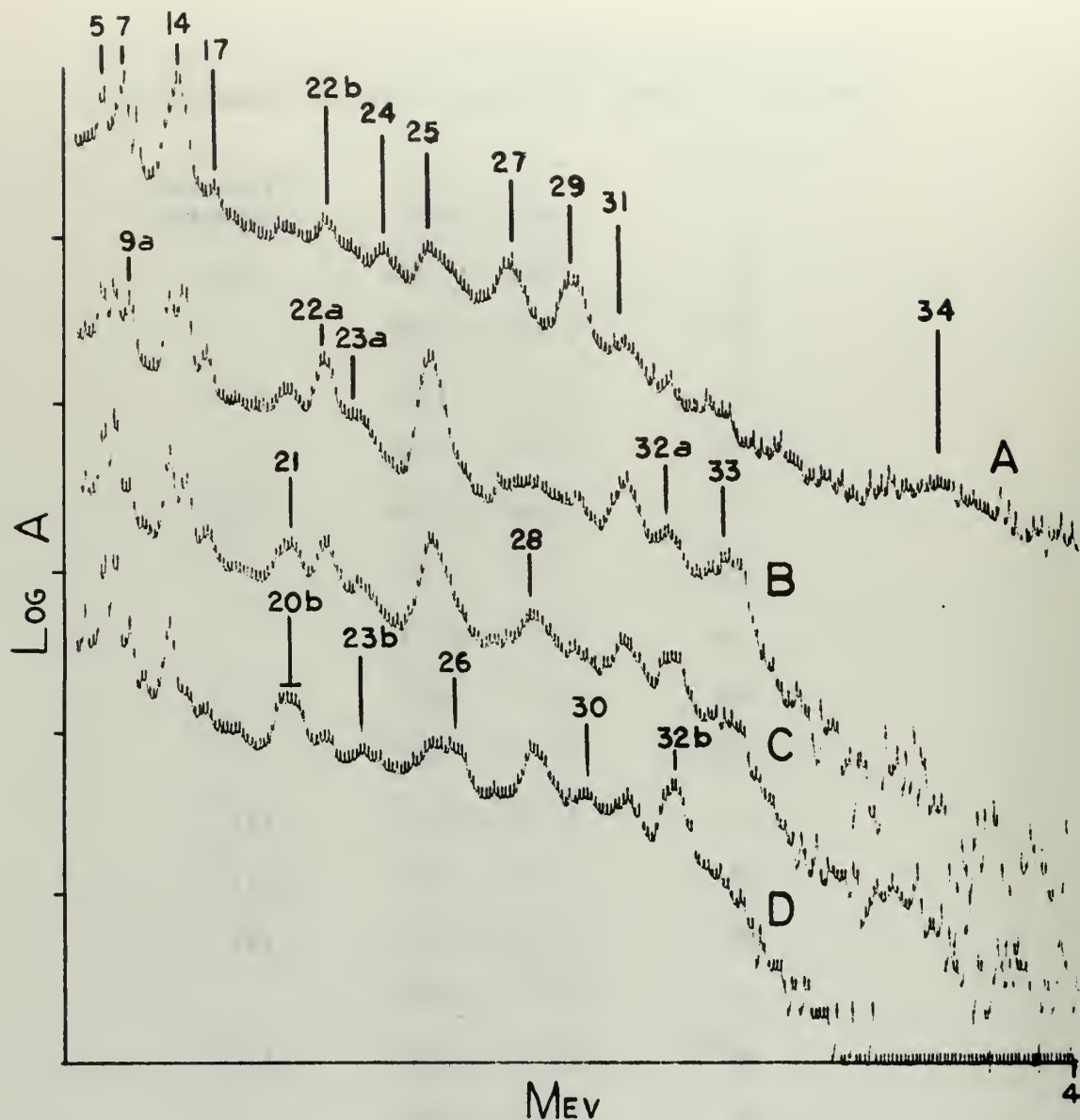
|     | Energy (Mev)      | Previously<br>Reported |
|-----|-------------------|------------------------|
| 1   | 0.012 $\pm 0.004$ |                        |
| 2   | 0.032 $\pm 0.002$ | (3)                    |
| 3   | 0.051 $\pm 0.002$ |                        |
| 4   | 0.079 $\pm 0.002$ | (2), (3)               |
| 5   | 0.148 $\pm 0.005$ | (3)                    |
| 6   | 0.161 $\pm 0.005$ | (3)                    |
| 7   | 0.201 $\pm 0.005$ |                        |
| 8   | 0.233 $\pm 0.005$ |                        |
| 9a  | 0.253 $\pm 0.005$ | (2), (3)               |
| 9b  | 0.265 $\pm 0.005$ |                        |
| 10  | 0.284 $\pm 0.005$ |                        |
| 11  | 0.310 $\pm 0.005$ | (3)                    |
| 12  | 0.364 $\pm 0.005$ |                        |
| 13a | 0.410 $\pm 0.005$ | (3)                    |
| 13b | 0.418 $\pm 0.005$ |                        |
| 14  | 0.439 $\pm 0.005$ |                        |
| 15  | 0.468 $\pm 0.005$ |                        |
| 16  | 0.527 $\pm 0.005$ |                        |
| 17  | 0.548 $\pm 0.005$ |                        |

Table I

# Photopeaks Present in Reactor Fuel Gas Gamma Spectra

|     | Energy (Mev)      | Previously<br>Reported |
|-----|-------------------|------------------------|
| 18  | 0.608 $\pm 0.005$ | (2)                    |
| 19  | 0.638 $\pm 0.005$ |                        |
| 20a | 0.81 -0.92        |                        |
| 20b | 0.83 -0.90        |                        |
| 21  | 0.886 $\pm 0.005$ | (3)                    |
| 22a | 1.01 $\pm 0.01$   |                        |
| 22b | 1.03 $\pm 0.01$   |                        |
| 23a | 1.14 $\pm 0.01$   |                        |
| 23b | 1.17 $\pm 0.01$   |                        |
| 24  | 1.23 $\pm 0.01$   | (3)                    |
| 25  | 1.44 $\pm 0.01$   | (3)                    |
| 26  | 1.58 $\pm 0.01$   | (3)                    |
| 27  | 1.74 $\pm 0.01$   |                        |
| 28  | 1.85 $\pm 0.01$   | (3)                    |
| 29  | 2.00 $\pm 0.01$   |                        |
| 30  | 2.05 $\pm 0.01$   |                        |
| 31  | 2.22 $\pm 0.01$   |                        |
| 32a | 2.37 $\pm 0.01$   |                        |
| 32b | 2.40 $\pm 0.01$   | (3)                    |
| 33  | 2.59 $\pm 0.01$   |                        |
| 34  | 3.3 $\pm 0.2$     |                        |

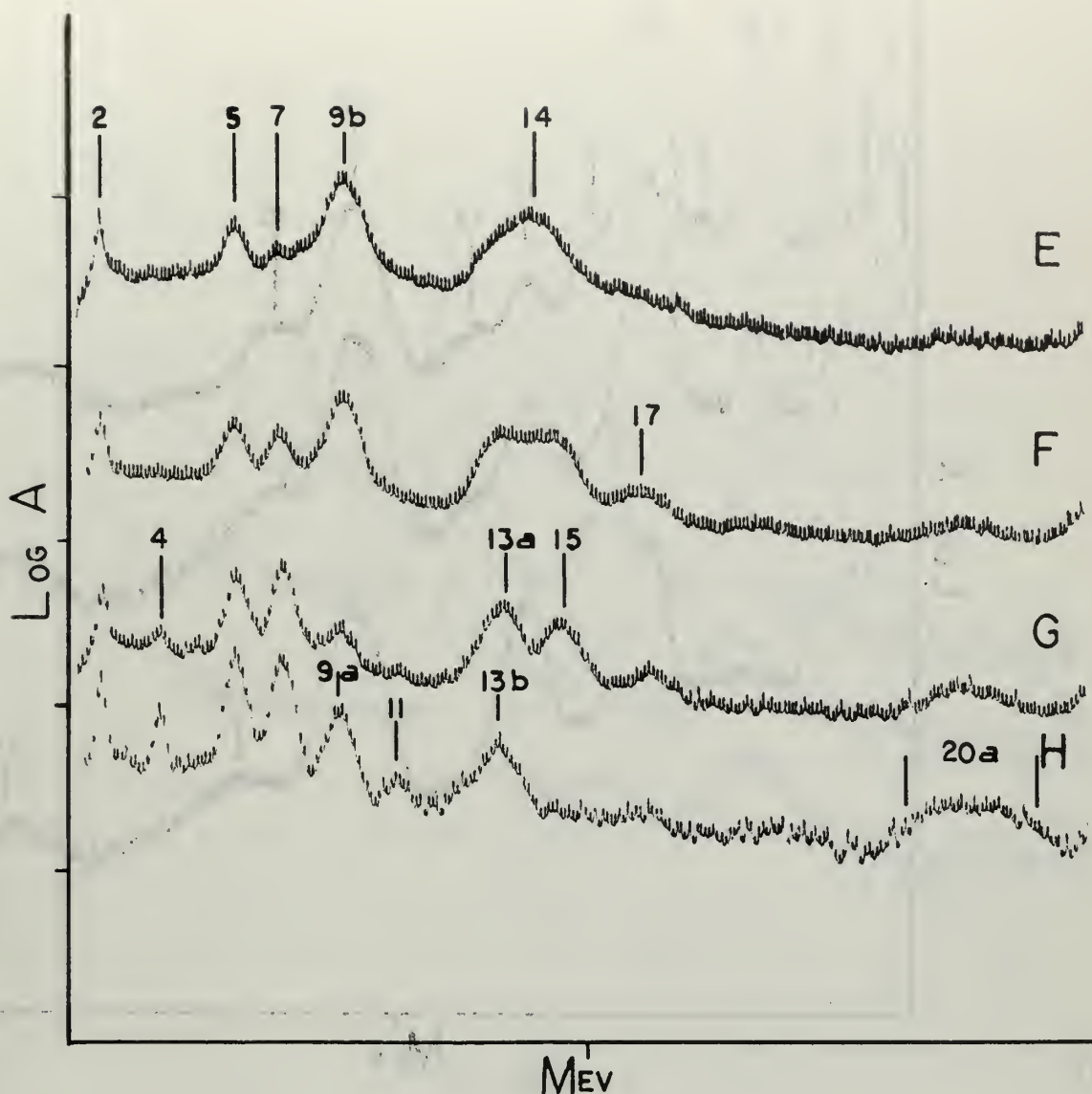
Table I (continued)



| SPECTRUM | RUN | COOLING TIME   | COUNTING TIME |
|----------|-----|----------------|---------------|
| A        | 16  | 6.40 minutes   | 1 minute      |
| B        | 16  | 72.50 minutes  | 4 minutes     |
| C        | 16  | 146.60 minutes | 10 minutes    |
| D        | 16  | 248.00 minutes | 20 minutes    |

Early 4 Mev Spectra

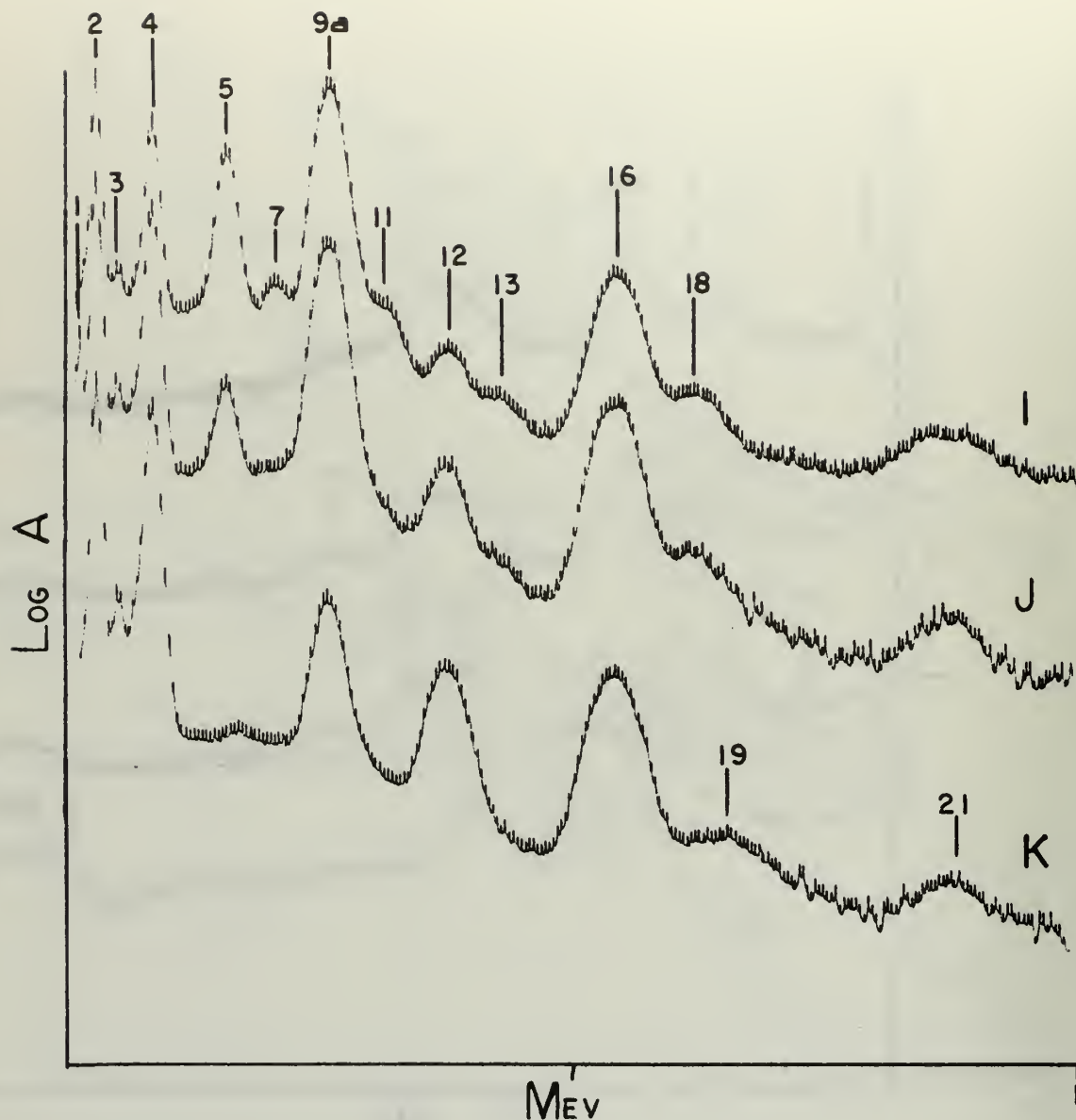
Figure 2



| SPECTRUM | RUN | COOLING TIME   | COUNTING TIME |
|----------|-----|----------------|---------------|
| E        | 15  | 11.55 minutes  | 1 minute      |
| F        | 15  | 41.50 minutes  | 4 minutes     |
| G        | 15  | 117.08 minutes | 4 minutes     |
| H        | 15  | 282.75 minutes | 20 minutes    |

Early 1 Mev Spectra

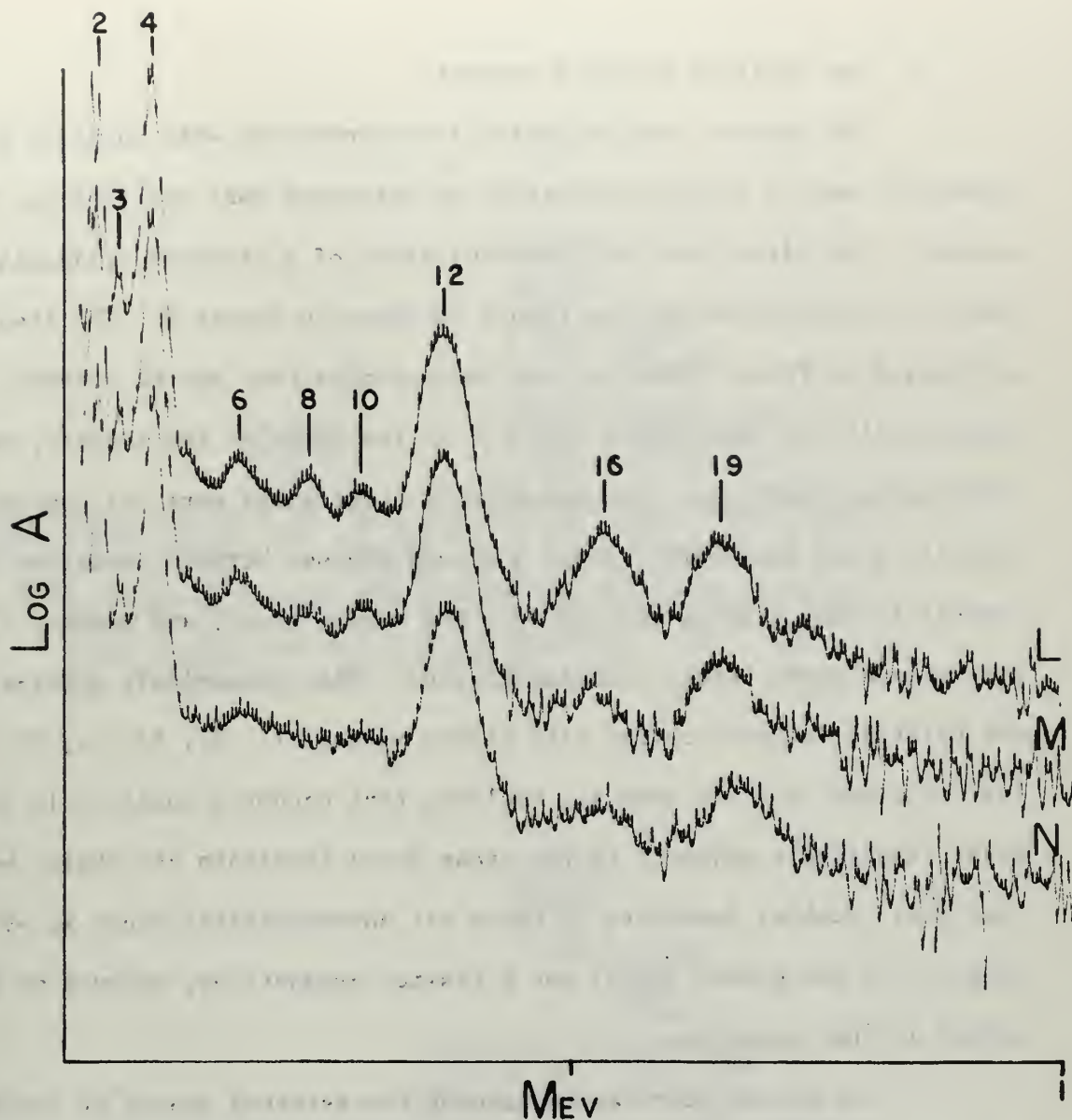
Figure 3



| SPECTRUM | RUN | COOLING TIME | COUNTING TIME |
|----------|-----|--------------|---------------|
| I        | 21  | 25.36 hours  | 40 minutes    |
| J        | 21  | 41.16 hours  | 40 minutes    |
| K        | 21  | 73.88 hours  | 100 minutes   |

Intermediate 1 Mev Spectra

Figure 4



| SPECTRUM | RUN | COOLING TIME | COUNTING TIME |
|----------|-----|--------------|---------------|
| L        | 21  | 7.149 days   | 1000 minutes  |
| M        | 21  | 12.033 days  | 1000 minutes  |
| N        | 21  | 21.700 days  | 1000 minutes  |

Late 1 Mev Spectra

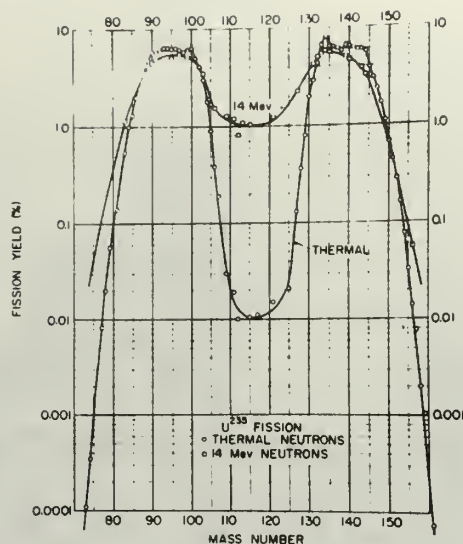
Figure 5

## B. The Nuclides Probably Present

The obvious starting point for determining what nuclides are present in such a complex mixture is to determine what nuclides may be present. The first step was a careful study of a standard triangular chart of the nuclides and the result is shown in Figure 6. The areas delineated in Figure 6 are all the decay chains that may be present, consistent with the constraints implicit in the scope of the project, and experimental technique. The two primary constraints were, of course, volatility and half life. About 1 minute elapsed between cessation of irradiation and drawing the gas into the sample ampul, and another 3 to 7 minutes elapsed before counting started. This effectively eliminated the volatile isobaric chains with atomic masses of: 92, 93, 94, 95, 141, 142, 143, and 144. In general, nuclides that occupy a position in the chart immediately adjacent to the areas shown terminate the chains because they are: stable; daughters to which all non-negligible decay is by beta (-) to the ground state; not a fission product; or, because no nuclide exists at that position.

The second step was to examine the selected groups of nuclides in order to estimate the relative abundances of each decay chain. There are three pertinent indicators for this. The first is the familiar plot of the Fission Yield curve for thermal neutrons as a function of A, such as the one in Figure 7, which indicates the relative production of each isobaric decay chain. This can further be refined, by noting the actual Fission Yields that have been measured experimentally, when they are available. An excellent compilation is the one by Katcoff (7). The third useful indicator is Independent Yield (8). This is an empirical parameter of each decay chain that gives an estimate of the relative yield of each member of the decay chain, and has been determined for atomic numbers in





Thermal Neutron Fission Yield (%) of  $U^{235}$  (7)

Figure 7

the ranges of 78 to 95 and 128 to 145. The Independent Yield is a function (a Gaussian curve) of the quantity  $Z - Z_p$ . A table of values of  $Z_p$  for applicable values of atomic mass is given in Table II. So for a nuclide of given atomic mass and atomic number, one determines  $Z - Z_p$ , and the IY is normally read from the curve. This can be a tedious process if many nuclides are being considered, so I have translated the graph into the more easily readable tabular form, and this is shown in Table III. Finally, I have summarized all the information from these three sources in Figure 8. The (m) in Figure 8 indicates the number applies to the metastable isomer. This information coupled with half life values shows that the following nuclides would be expected to constitute the most significant portion of the activity in the sample: Kr, A: 88, 89, 90; Br, A: 87, 88, 89; Xe, A: 135, 137, 138, 139, 140; I, A: 134, 135, 136, 137, 138. The presence of most of these nuclides was seen in the gamma spectra of the fuel gas.

$Z_p$  vs. A Values for the Thermal Fission of  $U^{235}$

| A  | $Z_p$ | A   | $Z_p$ |
|----|-------|-----|-------|
| 78 | 31.0  | 128 | 50.2  |
| 79 | 31.3  | 129 | 50.3  |
| 80 | 31.6  | 130 | 50.4  |
| 81 | 32.0  | 131 | 50.8  |
| 82 | 32.3  | 132 | 51.3  |
| 83 | 32.7  | 133 | 51.5  |
| 84 | 33.1  | 134 | 51.8  |
| 85 | 33.5  | 135 | 52.4  |
| 86 | 33.9  | 136 | 52.6  |
| 87 | 34.4  | 137 | 53.3  |
| 88 | 34.9  | 138 | 53.4  |
| 89 | 35.4  | 139 | 53.8  |
| 90 | 35.8  | 140 | 54.3  |
| 91 | 36.3  | 141 | 55.0  |
| 92 | 36.8  | 142 | 55.4  |
| 93 | 37.4  | 143 | 55.9  |
| 94 | 37.8  | 144 | 56.4  |
| 95 | 38.4  | 145 | 56.8  |
| 96 | 38.2  | 146 | 57.2  |
| 97 | 38.65 | 147 | 57.7  |
| 98 | 39.1  | 148 | 58.1  |
|    |       | 149 | 58.4  |
|    |       | 150 | 58.7  |

Table II

$Z - Z_p$  vs. Independent Yield (%)

| $Z - Z_p$ | Independent Yield | $Z - Z_p$ | Independent Yield |
|-----------|-------------------|-----------|-------------------|
| 0.0       | 58                | 1.5       | 6                 |
| 0.1       | 56                | 1.6       | 3.8               |
| 0.2       | 54                | 1.7       | 3.0               |
| 0.3       | 52                | 1.8       | 2.2               |
| 0.4       | 50                | 1.9       | 1.5               |
| 0.5       | 44                | 2.0       | 0.9               |
| 0.6       | 40                | 2.1       | 0.6               |
| 0.7       | 36                | 2.2       | 0.4               |
| 0.8       | 31                | 2.3       | 0.25              |
| 0.9       | 26                | 2.4       | 0.1               |
| 1.0       | 19                | 2.5       | 0.08              |
| 1.1       | 15                | 2.6       | 0.04              |
| 1.2       | 11.5              | 2.7       | 0.02              |
| 1.3       | 10                | 3.2       | 0.005             |
| 1.4       | 8                 |           |                   |

Table III

|     |            |     |                |     |             |     |      |     |     |
|-----|------------|-----|----------------|-----|-------------|-----|------|-----|-----|
| 131 | 2.93       | 133 | 6.62 m<br>0.08 | 135 | 6.3 m<br>38 | 137 | 138  | 139 | 140 |
|     |            |     |                |     |             | 6.0 | 5.49 | 54  | 38  |
|     |            |     |                |     |             | 36  | 40   | 54  | 52  |
| 131 | 3.1<br>0.4 | 132 | 30             | 133 | 6.9<br>6    | 135 | 137  | 138 | 139 |
|     |            |     |                |     |             | 3.1 | 52   |     | 31  |
|     |            |     |                |     |             | 50  |      |     |     |

X<sub>E</sub>

I

|    |         |    |      |    |     |    |      |     |      |
|----|---------|----|------|----|-----|----|------|-----|------|
| 83 | 0.544 m | 85 | 0.08 | 87 | 3.8 | 88 | 89   | 90  | 91   |
|    |         |    |      |    |     |    | 4.39 | 5.4 | 3.45 |
|    |         |    |      |    |     | 15 | 40   | 50  | 52   |
| 82 | 0.02    | 84 | 1.5  | 86 | 15  | 87 | 88   | 89  |      |
|    |         |    |      |    |     |    | 56   | 50  |      |

K<sub>R</sub>

B<sub>R</sub>

# FISSION YIELD-INDEPENDENT YIELD

FIGURE 8

#### IV. Photopeak Analysis

##### A. General

The gamma spectra obtained from the fuel gas were calibrated, in the standard manner, by superimposing on them the spectra of several useful standards. Those most frequently used were: Cs<sup>137</sup>: 32 Kev and 0.662 Mev; Au<sup>198</sup>: 69 Kev and 0.411 Mev; Mn<sup>54</sup>: 0.842; and Co<sup>60</sup>: 1.17, 1.33, and 2.50 Mev. The multichannel analyzer used did not seem to exhibit the linearity of energy per channel that it has been credited with (3), however, these nuclides provided sufficient points to determine the photopeaks in the fuel gas spectra to within a few Kev, with good assurance. The energy of each photopeak was calculated by measuring the distance along the energy scale from the nearest standard peak, using the energy per channel ratio of the nearest bracketing peaks.

Parenthetically, it should be noted that the standards used did not determine the peaks in the fuel gas spectra above 1 Mev with nearly as great a precision as those below 1 Mev. This was considered to be of marginal significance since the peaks in this region were not prominent enough to conduct a detailed half life analysis using the methods described below. As a rule, any of the nuclides of interest with gammas in the higher range also had peaks in the lower range, so there was sufficient information contained in the latter region of the spectra.

Now then, the energy calibration, coupled with the earlier analysis of the possible and probable nuclides, allows a correlation between the nuclides and the photopeaks. One other tool of analysis is available, namely half life determination.

##### B. Half Life Analysis

Several efforts were made to determine the short and intermediate component half lives of photopeaks in the following intervals: 0.14 - 1.17

Mev; 0.17 - 0.24 Mev; 0.24 - 0.29 Mev; 0.33 - 0.40 Mev; 0.40 - 0.48 Mev (the members of the trio were analyzed both collectively and individually); 0.49 - 0.57 Mev; and the photoplateau, 0.81 - 0.91 Mev. Referring to Figure 2 and Figure 4, these intervals correspond, respectively, to peaks: 5; 7; 9a and 9b; 12; 13a, 13b, 14, 15; 16; 20a. The half life determinations were made by following the decay of the photopeaks with a single channel analyzer which gave repetitive outputs of the integrated number of disintegrations under the photopeak per unit time. These data were analyzed using the computer program, FRANTIC II (9), which has been adapted for use on the NPS IBM 360 Duplex computer.

FRANTIC is a very capable and flexible program designed for analysis of radioactive decay problems and is widely used by chemists and physicists at this institution. There are several modes in which it can be used, and it was used in a mode in which data of the type just described are inputted, along with estimates of the component half lives. The program then uses a iterative least squares method to determine which of the half life estimates best fit the input data. The program can cope with photopeaks containing up to 10 components, however, to get acceptable results each component half life should differ from the one of nearest magnitude by about a factor of two.

FRANTIC was used to conduct about forty analyses of the energy intervals listed above. Typically, the program was used to look for one to six components during each analysis. The early results, with only a few exceptions, did not show the expected number of components. Usually, only one half could be found that agreed well with accepted literature values for the probable components. It was believed that results of this type were obtained for two reasons. The first is that the peaks of interest were too strongly influenced by adjacent peaks; this can be seen easily in

the spectra. A second complicating effect is that the single channel analyzer measures the portion of the Compton scatter distribution under the peak of interest for all peaks of higher energy. The effect was that these interferences masked the contributions of the minor components in the peak. So a technique was developed to remove these two interferences.

### C. The Two Analyzer Technique

#### (1) Description

The principle is to follow the activity decay of a small energy range on each side of the peak being analyzed. Then assume this general level of activity is constant between the two intervals, i.e., under the peak of interest, and subtract the appropriate proportionate amount from the activity of the photopeak. This is accomplished with two single channel analyzers in the following manner. After appropriate calibration, one analyzer is set with an energy window that includes the photopeak. The second analyzer is set on a wider energy window and slaved to the first analyzer so that it counts simultaneously with it. After the data are taken the counts per counting interval from the first scaler are subtracted, point for point, from the same data from the second scaler. The result is then proportioned to the energy interval of the first analyzer and subtracted, point for point, from the counts registered by the first analyzer.

The data manipulation can be expressed more simply algebraically. If:  $W_1$  = width of energy window for analyzer monitoring the peak;  $A_1$  = number of counts registered by first analyzer, and  $W_2$  and  $A_2$  are the corresponding quantities for the second analyzer; then the corrected FRANTIC input data point,  $P$ , equals:  $A_1 - \frac{W_1}{W_2 - W_1} (A_2 - A_1)$ . This is readily simplified to:  $P = C_1 A_1 - C_2 A_2$ , where  $C_2 = \frac{W_1}{W_2 - W_1}$  and  $C_1 = C_2 + 1$ . Then it requires only a simple modification to the computer program to input  $A_1$ ,  $A_2$ ,  $C_1$  and  $C_2$ , and have  $P$  computed for each data point and used in the calculations of the program.

Experience showed that it was necessary to determine  $C_1$  and  $C_2$  in a way that obviated the uncertainty in  $W_1$  and  $W_2$ , rather than calculating them directly from  $W_1$  and  $W_2$ . The constants can be determined experimentally by using a standard sample that has a constant level of Compton distribution across the ranges to be analyzed with the single channel analyzers.  $\text{Co}^{60}$  is ideal for windows below 1 Mev. If the analyzers are set with the desired windows, and  $C_1$  and  $C_2$  are correct, then when looking at a level spectrum, the corrected data point,  $P$ , must equal zero. So from the basic equation for  $P$ , we have  $C_2 = \frac{A_1}{A_2 - A_1}$ . So to determine  $C_1$  and  $C_2$  it is only necessary to make a few counts and calculate  $C_2$ . One important advantage with this method of determining  $C_1$  and  $C_2$  is that their precision is limited only by the length and number of counts one is willing to make to determine  $A_1$  and  $A_2$ .

The Two Analyzer Technique of data treatment was tested with two test runs and the results obtained from FRANTIC are listed in Table IV below. The test was to analyze the 0.85 Mev peak in a sample that contained:  $\text{Mg}^{27}$ ,  $t_{1/2} = 9.1$  minutes, photon energy = 0.835 Mev;  $\text{Mn}^{56}$ ,  $t_{1/2} = 2.58$  hours, photon energy = 0.845 Mev; and 5.1 minute  $\text{Cu}^{64}$  and 12.9 hr  $\text{Cu}^{66}$  whose principle spectral peaks are the 0.51 Mev and 1.02 Mev annihilation peaks. The Cu isotopes were added to provide a high level of Compton scatter distribution. The  $\text{Mg}^{27}$  did not activate sufficiently to be detected, but the Two Analyzer Technique succeeded very well in removing the contribution made to the peak by the Compton distribution of the higher energy peaks. The sample for the second run was intended to differ from the first only in that the amount of Mn was about one one-half of that in the sample used in the first run. However, the magnesium salt used in the sample of the second run contained an impurity that activated so well that it masked even the Cu activity. The unidentified impurity

had a nominal half life of a few hours and contributed the 35.2 minute half life shown in Table IV.

#### PHOTOPEAK ANALYSIS

|            | Photopeak Data<br>Only | Two Analyzer<br>Technique |
|------------|------------------------|---------------------------|
| Test Run 1 | 2.72 hr<br>5.1 min     | 2.66 hr                   |
| Test Run 2 | 2.74 hr<br>35.2 min    | 2.60 hr                   |

Table IV

#### (2) Application to Fuel Gas Spectrum

The Two Analyzer Technique was applied to the photopeaks listed at the beginning of Section IV B above and it gave improved results, in several cases. The results were improved in the same manner as that shown in the test runs, i.e., it eliminated spurious half lives that were contributed by the Compton distribution. However, it did not allow detection of additional minor component half lives in the photopeak, as had been hoped for. For a comparison of the results obtained from applying both methods to fuel gas spectra, see the entries in Table V below for Peak No. 7.

## V. Results

The heart of the experimental results has been displayed in the spectra of Figures 2-5. Because of the many peaks involved, and the even greater amount of nuclidic data that must be correlated, the interpretation of the spectra is done largely in tabular form below. The peak numbers listed in Table V correspond to the peak numbers given in Table I and Figures 2-5. The information on the nuclides was taken largely from the very convenient compilation by Crouthamel (10). A (a) after a half life value indicates it was determined using photopeak data only. A (b) indicates it was determined using the Two Analyzer Technique. A number in parenthesis represents a bibliography entry, as usual. Where several acceptable measurements were obtained all are listed. FRANTIC outputs a standard deviation for each half life determined, and this is not listed in order to conserve space, and because the FRANTIC result is not shown if the standard deviation was more than 5% of the half life literature value. Photon energy is stated in Mev. Where a nuclide assigned to a peak is enclosed in parenthesis, the parenthesis indicate that such an assignment is possible but not well founded. A list of the nuclides discovered is shown in Table VI.

# SUMMARY OF EXPERIMENTAL RESULTS

| Peak No. | Half Lives                                                                                         | Possible Nuclides                                                                                           | Nuclide Assignment/Remarks                                                                                                                                                                                                                              |
|----------|----------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| 1.       | See Figure 4                                                                                       | $Kr^{85}$<br>$11Y$<br>$Sr^{91}$<br>$9.7h$                                                                   | $Br^{82}$<br>$35.3h$<br>$Y^{91m}$<br>$50m$<br>Peak only displayed in Spectrum J, but it occurred in late spectra at least 6 different irradiations. It was deleted from Spectra H through L during printing.<br>$(Kr^{85}) (Br^{82}) (Sr^{91}: Y^{91})$ |
| 2.       | 21h growth (3)                                                                                     | All of Br and Kr nuclides and their daughter products.<br>See Figure 6.                                     | No growth seen in any spectra of this project.                                                                                                                                                                                                          |
| 3.       |                                                                                                    | $Xe^{133}$<br>$0.081, 0.053$<br>$5.28d$                                                                     | $Xe^{133}$                                                                                                                                                                                                                                              |
| 4.       | 5.29d (3)                                                                                          | $Xe^{133}$<br>$I^{131}$<br>$0.080$<br>$8.09d$                                                               | $Xe^{133}$                                                                                                                                                                                                                                              |
| 5, 6.    | 4.69h,<br>21.4h (a)<br>3.94h (a)<br>4.97h (b)<br>2.3h,<br>7.94h,<br>3.6m,<br>21.9m (a)<br>4.5h (3) | 5. $Kr^{85m}$<br>$0.150$<br>$4.36h$<br>$Xe^{135}$<br>$0.140$<br>$0.2h$<br>6. $Xe^{131}$<br>$0.164$<br>$12d$ | $Kr^{88}$<br>$0.166$<br>$2.77h$<br>$Kr^{85m}, Kr^{88}, (Xe^{135})$<br>$Xe^{131}$                                                                                                                                                                        |

Table V

# SUMMARY OF EXPERIMENTAL RESULTS

| Peak No. | Half Lives                                                                                                                       | Possible Nuclides                   |                                                                           | Nuclide Assignment/Remarks              |
|----------|----------------------------------------------------------------------------------------------------------------------------------|-------------------------------------|---------------------------------------------------------------------------|-----------------------------------------|
| 7.       | 25.9m,<br>1.2h,<br>3.1h (a)<br><br>2.51h (b)<br><br>3.0h (3)                                                                     | Kr <sup>88</sup><br>0.191<br>2.77h  | Kr <sup>89</sup><br>0.22<br>3.2m<br><br>I <sup>136</sup><br>0.210<br>1.4m | Kr <sup>88</sup>                        |
| 8.       | See Figure 5                                                                                                                     | Xe <sup>133m</sup><br>0.233<br>2.3d |                                                                           | Xe <sup>133m</sup>                      |
| 9a, b.   | 1.7m, 30.5m,<br>11.5h (a)<br><br>11.03h (a)<br><br>10.75h (a)<br><br>10.65h (a)<br><br>8.58h (b)<br><br>9.1h (3)<br><br>2.5d (3) | Xe <sup>135</sup><br>0.250<br>9.13  | I <sup>136</sup><br>0.270<br>1.4m                                         | Xe <sup>135</sup> , (I <sup>136</sup> ) |
| 10.      | See Figure 5                                                                                                                     | I <sup>131</sup><br>0.284<br>8.09d  | I <sup>136</sup><br>0.270<br>1.4m                                         | I <sup>131</sup>                        |
| 11.      | See Figure 4<br>4.2h (3)                                                                                                         | Kr <sup>85m</sup><br>0.305<br>4.36h | Ba <sup>140</sup><br>0.301<br>12.8d                                       | Kr <sup>85m</sup>                       |
| 12.      | See Figure 5                                                                                                                     | Xe <sup>135</sup><br>0.36<br>9.13   | I <sup>131</sup><br>0.364<br>8.09d                                        | I <sup>131</sup>                        |

Table V (continued)

# SUMMARY OF EXPERIMENTAL RESULTS

| Peak No. | Half Lives    | Possible Nuclides |                    | Nuclide Assignments/Remarks                              |
|----------|---------------|-------------------|--------------------|----------------------------------------------------------|
| 13a,     | 13, 14, 15:   | I <sup>136</sup>  | Kr <sup>87</sup>   | Kr <sup>87</sup> , Cs <sup>138</sup> , Xe <sup>138</sup> |
| 13b,     | 4.6m, 27.7m,  | 0.388             | 0.403              | (Br <sup>84m</sup> ) (Xe <sup>137</sup> )                |
| 14,      | 1.56h (a)     | 1.4m              | 1.3h               |                                                          |
| 15.      |               |                   |                    |                                                          |
|          | 13:34m, 1.7h, | Xe <sup>138</sup> | La <sup>140</sup>  |                                                          |
|          | 6.5h (a)      | 0.42              | 0.438              |                                                          |
|          |               | 17m               | 40.2h              |                                                          |
|          | 13:4.5h       |                   |                    |                                                          |
|          | 2.2h (3)      | Br <sup>84m</sup> | Cs <sup>138</sup>  |                                                          |
|          |               | 0.44              | 0.463              |                                                          |
|          |               | 6.0m              | 32.2m              |                                                          |
|          |               | Xe <sup>137</sup> | (5)                |                                                          |
|          |               | 0.44              |                    |                                                          |
|          |               | 4.2m              |                    |                                                          |
| 16.      | See Figure 5  | I <sup>133</sup>  | Xe <sup>135m</sup> | I <sup>131</sup> , (I <sup>133</sup> )                   |
|          |               | 0.53              | 0.53               |                                                          |
|          |               | 20.8h             | 15.6m              |                                                          |
|          |               | I <sup>131</sup>  |                    |                                                          |
|          |               | 0.51              |                    |                                                          |
|          |               | 8.09d             |                    |                                                          |
| 17.      | See Figure 2  | Sr <sup>91</sup>  | Y <sup>91m</sup>   | Cs <sup>138</sup>                                        |
|          |               | 9.7h              | 0.55               |                                                          |
|          |               |                   | 50.3m              |                                                          |
|          |               | Cs <sup>138</sup> | Br <sup>82</sup>   |                                                          |
|          |               | 0.55              | 0.554              |                                                          |
|          |               | 32 min            | 36h                |                                                          |
| 18.      | See Figure 4  | Xe <sup>135</sup> |                    | Xe <sup>135</sup>                                        |
|          |               | 0.61              |                    |                                                          |
|          |               | 9.13h             |                    |                                                          |
| 19.      | See Figure 5  | I <sup>131</sup>  | Sr <sup>91</sup>   | I <sup>131</sup>                                         |
|          |               | 0.637             | 0.645              |                                                          |
|          |               | 8.09d             | 9.7h               |                                                          |

Table V (continued)

# SUMMARY OF EXPERIMENTAL RESULTS

| Peak No. | Half Lives                   | Possible Nuclides |                   | Nuclide Assignments/Remarks               |
|----------|------------------------------|-------------------|-------------------|-------------------------------------------|
| 20a,     | 40.7m,                       | I <sup>132</sup>  | I <sup>133</sup>  |                                           |
| 20b,     | 2.91h (a)                    | 0.78              | 0.85              |                                           |
| 21.      |                              | 2.3h              | 21h               |                                           |
|          | 2.54h,                       |                   |                   |                                           |
|          | 2.68h (b)                    | I <sup>134</sup>  | Kr <sup>87</sup>  |                                           |
|          |                              | 0.85              | 0.85              |                                           |
|          | 2.7h (3)                     | 52.5m             | 76m               |                                           |
|          |                              | Kr <sup>88</sup>  | Br <sup>84m</sup> |                                           |
|          |                              | 0.85              | 0.88              |                                           |
|          |                              | 2.77h             | 6.0m              |                                           |
|          |                              | Y <sup>89m</sup>  | Rb <sup>88</sup>  |                                           |
|          |                              | 0.915             | 0.908             |                                           |
|          |                              | 50d               | 17.8m             |                                           |
|          |                              | Br <sup>84</sup>  | Sr <sup>91</sup>  |                                           |
|          |                              | 0.81              | 0.93              |                                           |
|          |                              | 32m               | 9.7h              |                                           |
| 22.      | About 20 minutes, initially. | Kr <sup>89</sup>  | Rb <sup>89</sup>  | (Kr <sup>89</sup> -Rb <sup>89</sup> ) and |
|          |                              | 3.2m              | 1.05              | (I <sup>135</sup> or Br <sup>84</sup> )   |
|          |                              |                   | 15.4m             |                                           |
|          |                              | I <sup>135</sup>  | Br <sup>84</sup>  |                                           |
|          |                              | 1.04              | 1.01              |                                           |
|          |                              | 6.68h             | 32m               |                                           |
| 23a,     |                              | I <sup>134</sup>  | I <sup>135</sup>  | (I <sup>134</sup> or I <sup>135</sup> )   |
| 23b.     |                              | 1.10              | 1.14              |                                           |
|          |                              | 52.5m             | 6.68h             |                                           |
| 24.      | 2.7h (3)                     | Kr <sup>89</sup>  | Rb <sup>89</sup>  | (Kr <sup>89</sup> -Rb <sup>89</sup> )     |
|          |                              | 3.2m              | 1.26              |                                           |
|          |                              |                   | 15.4m             |                                           |

Table V (continued)

# SUMMARY OF EXPERIMENTAL RESULTS

| Peak No. | Half Lives                   | Possible Nuclides                                                                   | Nuclides Assignments/Remarks                                             |
|----------|------------------------------|-------------------------------------------------------------------------------------|--------------------------------------------------------------------------|
| 25.      |                              | $I^{132}$<br>1.4<br>2.26h<br>$Cs^{138}$<br>1.43<br>32.2m<br>$Br^{84}$<br>1.46<br>6m | $I^{133}$<br>1.4<br>21h<br>$Sr^{91}$<br>1.4<br>9.7h                      |
| 26.      | 2.7h (3)                     | $Kr^{88}$<br>1.55<br>2.8h<br>$La^{140}$<br>1.60<br>40.2h                            | $Rb^{89}$<br>1.55<br>15m<br>$Kr^{88}$                                    |
| 27, 28.  | 2.7h (3)                     | $I^{135}$<br>1.72<br>6.68h<br>$Rb^{88}$<br>1.85<br>17.8m                            | $Kr^{88}$<br>2.87h<br>$Kr^{87}$<br>1.75<br>1.3h<br>$Kr^{88}$ ; $Rb^{88}$ |
| 29.      | 20 minutes;<br>See Figure 2. | $Xe^{138}$<br>2.01<br>17m                                                           | $Xe^{138}$                                                               |
| 30.      |                              | $Kr^{87}$<br>2.05<br>1.3h                                                           | $Kr^{87}$                                                                |

Table V (continued)

# SUMMARY OF EXPERIMENTAL RESULTS

| Peak No.     | Half Lives                   | Possible Nuclides                      |                                            | Nuclides Assignments/Remarks             |
|--------------|------------------------------|----------------------------------------|--------------------------------------------|------------------------------------------|
| 31           |                              | Xe <sup>138</sup><br>2.2<br>17m        | Cs <sup>138</sup><br>2.21<br>32.2m         | Xe <sup>138</sup> , Cs <sup>138</sup>    |
|              |                              | Kr <sup>89</sup><br>3.2m               | Rb <sup>89</sup><br>2.20<br>15.4m          |                                          |
| 32a,<br>32b. | 2.9h (3)                     | Kr <sup>88</sup><br>2.40<br>2.77h      | Br <sup>84</sup><br>2.47<br>32m            | Kr <sup>88</sup> (Br <sup>84</sup> )     |
| 33.          |                              | Kr <sup>87</sup><br>2.57<br>1.3h       | Rb <sup>89</sup><br>2.59<br>15.4m          | (Rb <sup>89</sup> ) (Cs <sup>138</sup> ) |
|              |                              | Cs <sup>138</sup><br>2.63<br>32.2m     |                                            |                                          |
| 34.          | Very short.<br>See Figure 1. | Rb <sup>89</sup><br>3.0,<br>3.5<br>15m | Br <sup>84</sup><br>3.03,<br>3.91<br>32min |                                          |

Table V (continued)

## NUCLIDES IDENTIFIED

### Volatile Nuclides:

$I^{131}$   
 $Xe^{131}$ ,  $Xe^{133m}$ ,  $Xe^{133}$ ,  $Xe^{135}$ ,  $Xe^{138}$   
 $Kr^{85m}$ ,  $Kr^{81}$ ,  $Kr^{88}$

### Active Daughters:

$Cs^{138}$   
 $Rb^{88}$

### Nuclides Probably Present:

$I^{133}$  (11),  $I^{134}$ ,  $I^{135}$  (11),  $I^{136}$   
 $Xe^{137}$   
 $Kr^{88}$ ,  $Kr^{89}$   
 $Br^{84m}$ ,  $Br^{84}$   
 $Rb^{89}$

Table VI

## VI. Conclusion

The dramatic increase in the complexity of the fuel gas spectra over earlier core gas spectra, and the concomitant increase in the amount of nuclidic information contained therein, certainly vindicated the new method of obtaining the gas.

The more complex spectra, however, require new and more capable automated and computerized means of half life determination, if all of the information available in the new spectra is to be retrieved. One approach that should be considered is to conduct a detailed mathematical analysis of the various equilibria occurring between active parents and active daughters. Alternatively, the gases could be separated rapidly prior to applying gamma-ray spectrometry.

The Two Analyzer Technique of half life determination is a valuable improvement in the former direction, but it apparently was not sufficiently capable to cope with the fuel gas spectrum. The latter route has been tried (11) in work carried on concurrently with this project, and the fuel gas was fractionated chromatographically into iodine, krypton, and xenon fractions. The resulting spectra were much less complex, but the process, in its current state of development, is so slow that short lived nuclides cannot be detected.

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## Appendix I

### EQUIPMENT

1. Counting Chamber: A 2-inch thick lead cube, 22 inches square by 28 inches high, lined with 1/8-inch aluminum and 1/4-inch high copper content brass.
2. Scintillation Detector: Harshaw 12SW12-W3, 3-inch by 3-inch cylindrical well type NaI (Th) crystal with matched photomultiplier tube.
3. 512 channel multi-channel analyzer, consisting of the following components:
  - (a) Nuclear Data, Inc ND-180F Analog to Digital Converter
  - (b) Nuclear Data, Inc ND-180M 512 Channel Memory Unit
  - (c) Nuclear Data, Inc ND-180R Read out Control Unit
  - (d) Textronix, Inc, Rm 503 Oscilloscope
  - (e) Tally Corp, Model 1506 Reader/Perforator
  - (f) Teletype Corp, Teletypewriter
  - (g) Hewlitt-Packard, Co., Mosley 7590 CR Automatic Plotting System
4. Single channel analyzer (2 ea): Hewlitt-Packard Model 5201L Scaler-Timer
5. Rapid digital printer (2 ea): Hewlitt-Packard, Co., 562 A Digital Recorder.

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| 13. ABSTRACT                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                             |  |                                                                             |                 |
| <p>The volatile radioactive species produced by irradiation of AGN - 201 Reactor fuel elements were obtained by irradiation of the fuel in an evacuated inert container. The following 11 constituent nuclides, including two radioactive daughters, were identified by gamma ray spectrometry: <math>I^{131}</math>, <math>Xe^{131}</math>, <math>Xe^{131m}</math>, <math>Xe^{135}</math>, <math>Xe^{138}</math>, <math>Kr^{87}</math>, <math>Kr^{88}</math>, <math>Cs^{138}</math>, and <math>Rb^{88}</math>. Additionally, the possible presence of the following species was indicated: <math>I^{133}</math>, <math>I^{134}</math>, <math>I^{135}</math>, <math>I^{136}</math>, <math>Xe^{137}</math>, <math>Kr^{88}</math>, <math>K^{89}</math>, and <math>Pb^{89}</math>. The spectra studied were taken with a NaI (Th) crystal and they contained 34 photopeaks. An automated computerized method for half life analysis of complex photopeaks is described.</p> |  |                                                                             |                 |

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### KEY WORDS

LINK A

LINK B

LINE C

ROLE

WT

ROLE

WT

ROLE

VI

## ANALYSIS

CORE

FUEL

## GAMMA-RAY

## GAS

REACTOR

SPECTROMETRY



















































































































































































































































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